

A Systematic Review of Evidence and Implications of Spatial and Seasonal Variations of Volatile Organic Compounds (VOC) in Indoor Human Environments

Inês Paciência^{a,b,c}, Joana Madureira ^a, João Rufo ^{a,b,c}, André Moreira^{b,c}, and Eduardo de Oliveira Fernandes^a

^aInstitute of Science and Innovation on Mechanical Engineering and Industrial Management, Porto, Portugal; ^bFaculty of Medicine, University of Porto, Porto, Portugal; ^cPortugal & Centro Hospitalar São João, Porto, Portugal

ABSTRACT

Many volatile organic compounds (VOC) are classified as known or possible human carcinogens, irritants, and toxicants, and VOC exposure has been associated with asthma and other respiratory symptoms/diseases. This review summarizes recent quantitative data regarding VOC in four categories of indoor environments (schools, housing, offices, and other indoor) and compares the types and concentration levels of individual VOC that were detected, measured, and reported according to season (cold and warm). The influence of outdoor air on concentrations of indoor VOC was also assessed as ratios of indoor versus outdoor. Papers published from 2000 onward were reviewed and 1383 potentially relevant studies were identified. From these, 177 were removed after duplication, 1176 were excluded for not meeting the review criteria, and 40 were included in this review. On average, higher mean concentrations of indoor VOC were found in housing environments, in offices, and in the cold season. Volatile organic compounds are commonly present in indoor air and specific compounds, and their concentrations vary among indoor environments and seasons, indicating corresponding differences in sources (indoors and outdoors). Actions and policies to reduce VOC exposures, such as improved product labeling and consumer education, are recommended.

Many volatile organic compounds (VOC) are classified as known or possible human carcinogens, irritants, and toxicants (U.S. Environmental Protection Agency, 2012a). In addition, several studies linked VOC exposure to asthma and other respiratory symptoms/diseases (Nurmatov et al. 2015). Valcke and Haddad (2015) demonstrated that human responsiveness to VOC not only was dependent upon chemical concentrations but also needed to consider sensitive subpopulations such as neonates, toddlers, and pregnant women. It is widely recognized that indoor concentrations of many VOC are higher than levels in outdoor air (Annesi-Maesano et al. 2013; Edwards et al. 2001b; Guo et al. 2004; Madureira et al. 2009; 2015a, 2015b). Most exposures to VOC occur indoors, as a result of numerous indoor emission sources, low ventilation rates, and time spent indoors. Important indoor VOC sources include construction materials, furnishing, paints, glues,

heating appliances, tobacco smoke, cooking, cleaning products, and pesticides (Annesi-Maesano et al. 2013; Edwards et al. 2001a; Jurvelin 2003; Zhang et al. 2002). Outdoor air also contributes to indoor VOC levels, mainly because of their transport to indoors such as vehicle emissions and proximity to industrial activities the major ambient sources (U.S. Environmental Protection Agency, 2012b; Miller, Xu, and Luginaah 2009).

Based on health outcomes, the wide variety of sources including indoors and outdoors, the type and levels of VOC detected in different indoor environments, and time spent indoors highlight the need to summarize and compare data on VOC types and their levels in various microenvironments within and between indoor and outdoor air, and consequently, enhance available information on exposure to VOC indoors. Beyond the spatial variations (indoor/outdoor) and differences related to each microenvironment, such as homes, schools, offices, hospitals, and

CONTACT Joana Madureira  jmadureira@inegi.up.pt  Institute of Science and Innovation on Mechanical Engineering and Industrial Management, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal.

Authors Inês Paciência and Joana Madureira contributed equally to the work.

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stores, the season when samplings occur also influences the concentration of VOC indoors. Several studies demonstrated a significant seasonal cycle (Kim, Chun, and Jo 2015; Pegas et al. 2011b; Rehwagen, Schlink, and Herbarth 2003), probably due to (1) lower mobility of air in the wintertime, (2) longer persistence of the compounds due to reduced photochemical degradation (Fuselli et al. 2010), and (3) gradual accumulation (Pegas et al. 2011b). The large number of VOC combined with the time that people spend indoors justifies considering indoor air pollution among one of the most significant environmental health risks and emphasizes the importance of characterization of different indoor environments in order to understand the contribution of each on human exposure to VOC.

The purpose of this study was to review, summarize, and compare existing quantitative data concerning spatial (four major human environments: schools, housing, offices, and other indoor) and seasonal variation (cold and warm) of VOC concentrations that were detected, measured, and reported both indoors and outdoors. These data may be useful to many investigators and stakeholders interested in this environmental topic, namely, those responsible for adopting appropriate risk management strategies.

Methods

Search Strategy

Following the 2009 “Preferred Reporting Items for Systematic Reviews and Meta-Analyses” (PRISMA) guidelines (Moher et al. 2015), original published studies from 2000 to 2015 were searched through PubMed, Scopus, and Web of Science. The search included the combined terms “volatile organic compounds AND indoor air AND outdoor air.” Additionally, a cross-reference check was performed to search for additional potential studies. All studies that fulfilled the following inclusion criteria were considered: published in English; VOC measured in both indoor and outdoor environments; and VOC measured in at least one season. The exclusion criteria were studies reporting modeled or estimated exposure.

Data extraction

Two researchers independently screened titles and abstracts of identified studies, and reviewed the full text of studies meeting the inclusion and exclusion criteria. Disagreements were resolved through discussion between the two researchers. For each publication, the compounds and collected information on VOC concentration by outdoor and indoor environments, season, and data on exposure to tobacco smoke were identified.

Definitions and analysis

Studies that reported mean concentration values for different VOC were considered in data analyses, which was stratified by space (outdoor and indoor environments) and season (cold—winter and fall; warm—summer and spring). All studies that defined the sampling period month(s) and the season (winter, spring, summer and/or fall) or season periods (cold and/or warm) and whose results were presented in accordance with this definition were included in this study and categorized according to the preceding definition—warm and cold season.

Indoor environments comprised four categories: (1) schools, (2) housing such as homes and hotels, (3) offices, and (4) other indoor. This last category included shopping malls, hospitals and dental clinics, restaurants, photocopy centers, and museums. All eligible studies are described in the supplementary file stratified by space and season and with the information on exposure to tobacco smoke, when applicable.

No restrictions were made in the selection of studies according to analytical method that were used for identification and quantification of VOC. In the included studies, the most reported method was gas chromatography coupled with mass spectrometry (GC-MS), which was used in 18 studies, followed by gas chromatography coupled with flame ionization detector (GC-FID) (11 studies), whereas only 6 studies used gas chromatography coupled with mass selective detector (GC-MSD) and only 1 used gas chromatography coupled with an electron capture detector (GC-ECD). Some studies (three) reported the use of high-performance liquid chromatography (HPLC).

Distribution of quantitative variables was determined using the Shapiro-Wilk test, and the Mann-Whitney or Kruskal-Wallis test was used to test comparisons between VOC concentrations, indoor and outdoor, and seasons for the four indoor environments. All of the analyses were performed using the Statistical Package for the Social Sciences (SPSS), version 17.0, and the level of statistical significance was set at .05.

Results

Individual VOC in different indoor environments

Initially, 1393 papers were retrieved from the search, including overlapping publications across the three databases. After removal of duplicates (177), 1123 papers were excluded by title and abstract screening. Another 53 papers were removed after assessment of the full text, as these did not meet inclusion criteria (Table 1). Figure 1 shows the number of studies identified and included/excluded. Ultimately, 40 papers were included in this review and are described in detail in the supplementary file. Twelve studies were conducted at schools, 21 in housing environments,

4 in offices, and 7 in other indoor environments, between 1996 and 2014 (Table 2).

At schools, 33 VOC were measured and reported, of which 18 were measured in both seasons, with 12 exclusively in the warm and only 3 exclusively in the cold season. Higher mean concentrations for specific VOC, with measurements in both seasons (10 of 18), were found during the cold season (Figure 2). The concentrations ranged between nondetectable and 160 µg/m³; the highest level was noted for *m/p*-xylene and the lowest for various specific VOC with both values during cold season (supplementary file). A greater number of specific VOC were identified in housing environments (81), mostly measured in both seasons (74), with 6 measured in the warm and 1 in the cold season (tetrachlorocarbon).

In contrast to schools, the highest mean concentrations for specific VOC were identified in the warm season; however, the highest levels detected in the cold season were for toluene (293 µg/m³) (Figure 3). The concentrations ranged between non-detectable and 293 µg/m³, being higher for toluene (cold season) and lower for various specific VOC (supplementary file).

In offices, 20 VOC were measured in both seasons (14), with 4 in the cold and the remainder in the warm season (2). Similar to housing environments,

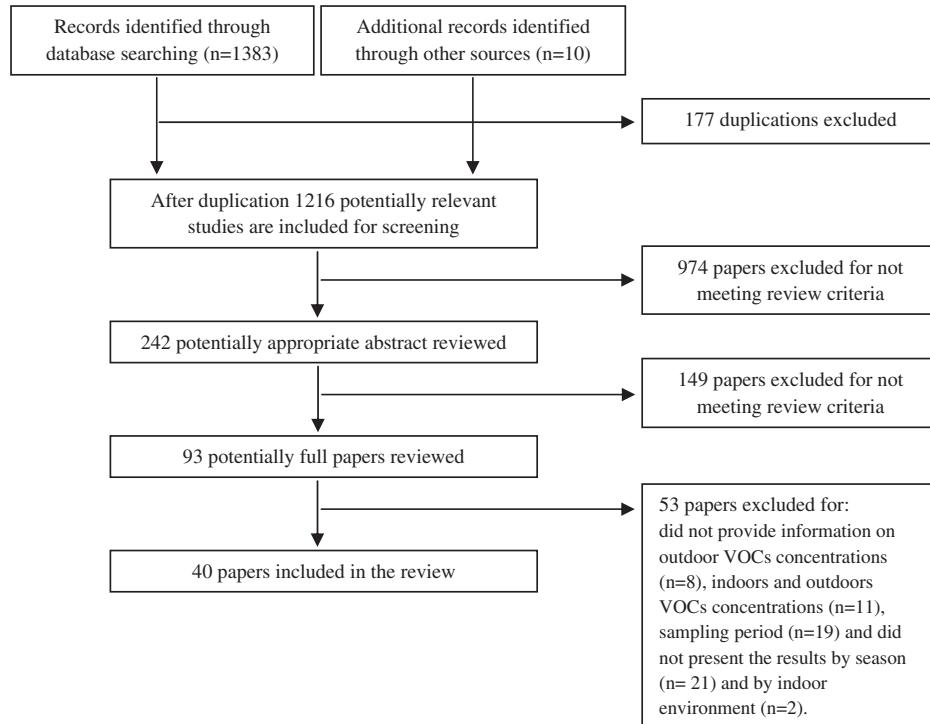


Figure 1. Systematic review flowchart.

Table 1. Excluded studies.

Reference	Year	Country	Indoor environment (n)	Season	Analytical method	Reasons for exclusion
Batterman et al., 2006	–	USA	Homes (1)	–	GC-MS	Did not provide information about sampling period
Batterman et al., 2007	–	USA	Homes (15)	–	GC-MS	
Caselli et al., 2009	–	Italy	Newspaper stands (16)	–	GC-MS	
de Gennaro et al., 2013	–	Italy	8 schools	–	TD-GC-MS	
Elkilani and Bouhamra, 2001	–	Kuwait	Homes (10)	–	GC	
Geiss et al., 2011	–	Italy	Car showrooms	–	GC-FID	
Guo et al., 2003	–	Hong Kong	Restaurants (4), schools (6), offices (6), shopping malls (6) and homes (6)	–	GC-MS	
Lerner et al., 2012	–	Argentina	Enterprise (laboratories, sewing workrooms, electromechanical repair and car painting centers, food shops and photocopy center)	–	GC-FID	
Pandit et al., 2001	–	India	Homes (3)	–	GC-FID	
Phillips et al., 2005	–	USA	Homes (42)	–	GC-MS	
Shin and Jo, 2013	–	Korea	Homes (25)	–	GC-MS	
Shin and Jo, 2014	–	Korea	Homes (107)	–	GC-FID GC-MS	
Srivastava et al., 2000	–	India	Offices (1)	–	GC-FID	
Yoon et al., 2011	–	Korea	Schools (17)	–	GC-MSD	
Yoon et al., 2011	–	Korea	17 preschools	–	GC-MSD GC-NPD	
Batterman et al., 2012	2004, 2005, 2010	USA	Homes (288)	Winter, summer, fall, spring	GC-MS	Did not provide information by season
Canha et al., 2015	2010	France	Schools (17)	–	GC-FID	
Cheng et al., 2015	2008, 2009	Australia	Homes (40)	–	GC-MS GC-FID HPLC	
Du et al., 2015	2011-2012	USA	Homes (74)	–	GC-MS	
Duan et al., 2014	2011, 2012	China	50 homes	Heating and non-heating season	GC-MSD GC-FID	
Edwards et al., 2001	1996-1997	Finland	201 microenvironments: homes and workplace	Fall 1996 to winter 1997	GC-FID GC-MS	
Eklund et al., 2008	2003-2005	USA	Shopping center (1)	–	GC-MS	
Fuselli et al., 2010	2007-2009	Italy	Offices (1)	–	GC-MS	
Gallego et al., 2008	2000, 2001	Spain	7 public buildings; 54 homes	Spring-summer and winter of 2000; and summer and winter of 2001	GC-FID	
Hinwood et al., 2006	2000	Australia	27 microenvironments: homes, offices, restaurants and nightclubs	–	GC-MS	
Jia et al., 2010	2005, 2006	USA	10 commercial or industrial buildings	–	GC-MS	
Jia et al., 2012	2004, 2005	USA	Homes (162)	Summer, winter, fall and spring	GC-MS	
Jung et al., 2011	2006, 2007	USA	Homes (15)	Summer and winter	GC-MS	
Lee et al., 2002a	–	Hong Kong	Restaurants (4), schools (10), offices (10), shopping malls (9), homes (6)	–	GC-MS	

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Table 1. (Continued).

Reference	Year	Country	Indoor environment (n)	Season	Analytical method	Reasons for exclusion
Lu et al., 2007	2003-2004	Taiwan	Offices (86)	–	Photo-ionization detector (PID) for real-time monitoring of VOCs	
Mishra et al., 2015	2010-2012	Australia	25 schools	–	GC-MS	
Payne-Sturges et al., 2004	2000-2001	USA	Homes (37)	–	GC-MS	
Rehwagen et al., 2003	1994-2001	Germany	Homes (1499 indoor and 222 outdoor measurements)	–	GC-FID/ECD	
Sax et al., 2006	1999, 2000	USA	Homes (121)	Winter, summer, fall	GC-MSD	
Serrano-Trespalacios et al., 2004	1998, 1999	Mexico	Homes (30)	–	GC-MS	
Sexton et al., 2004	1999	USA	Homes	Spring, summer and fall	GC-MS	
Shinohara et al., 2013	2011-2012	Japan	Homes (19)	–	GC-MS	
Topp et al., 2004	1995-1997, 1996-1998	Germany	Homes (631)	–	GC-FID	
Chin et al., 2014	2010	USA	Homes (126)	Spring, summer, fall and winter	GC-MS	Did not provide information on outdoor concentrations
Delgado-Saborit et al., 2011	2005-2007	UK	Homes (155), workplaces (40)	–	GC-MS	
Jo and Kim, 2010	2009	Korea	Schools (1)	Summer	GC-MS	
Sohn et al., 2009	2004	Korea	Schools (55)	Summer, autumn and winter	GC-FID	
de Blas et al., 2012	2008	Spain	1 school	Winter	GC-MS GC-FID	Did not provide information by indoor and outdoor
Dodson et al., 2009	2000, 2004, 2005	USA	Homes (95)	–	GC-MS HPLC	
Massolo et al., 2010	2000-2002	Argentine	Kindergartens (92) and homes (92)	Winter	GC-MS	Did not provide information by indoor environment
Sakai et al., 2009	2004-2007	Japan	67 rooms from 56 buildings [Offices (53), shops (7), classrooms (3), library (2), conference hall (2)]	Summer and winter	GC-MS	
Walgraeve et al., 2011	2007-2009	Belgium	Homes (6)	–	GC-MS	Did not provide information by indoor and outdoor Did not provide information by season
Tham et al., 2004	–	Singapore	Offices (1)	–	GC	Did not provide information on outdoor concentrations Did not provide information about sampling period

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Table 1. (Continued).

Reference	Year	Country	Indoor environment (n)	Season	Analytical method	Reasons for exclusion
Ongwandee et al., 2011	2009	Thailand	Offices (17)	–	GC-MS	Did not provide quantitative results by indoor and outdoor
Sexton et al., 2007	1999	USA	70 microenvironments: homes, work/ school, other locations (grocery, stores, restaurants, shopping malls)	Spring, summer and fall	GC-MS	Did not provide information by season
Xiong et al., 2015	2003- 2007	USA	Homes (17)	–	GC-MS	Did not provide information by season

GC-MS: Gas chromatography coupled with mass spectrometry; GC-FID: Gas chromatography coupled with flame ionization detector; GC-MSD: Gas chromatography coupled with mass selective detector; GC-ECD: Gas chromatography coupled with an electron capture detector; GC-NPD: Gas chromatography coupled with nitrogen phosphorus detector; TD-GC-MS: thermal desorption-gas chromatography coupled with mass spectrometry

Table 2. Included studies.

Reference	Year	Country	Indoor environment (n)	Season	Analytical method	Seasonal variation
Adgate et al., 2004a	2000	USA	Homes (113) and schools (113)	Winter and spring	GC-MS	In schools and in homes, most VOCs were more frequently detectable and at higher concentrations in winter compared with spring
Adgate et al., 2004b	1997	USA	Homes (284)	Warm season	GC-MS	–
Bae et al., 2004	2001-2002	Korea	Shopping malls (32)	Cold season	GC-MS	–
Baez et al., 2003	1996, 1997, 1998	Mexico	Homes (2), Museums (2) and Offices (2)	Warm and cold season	HPLC	–
Chan et al., 2009	2006	China	Hotels (8)	Winter	GC-MS	–
Feng et al., 2004	2002	China	Hotels (4)	Warm season	HPLC	–
Fischer et al., 2000	1995	Netherlands	Homes ()	Cold season	GC	–
Godoi et al., 2009	2005	Brazil	Schools (2)	Cold season	GC-MS	–
Godwin and Batterman, 2007	2003	USA	Schools (29)	Warm season	GC-MS	–
Gokhale et al., 2008	2005	Germany	Homes (7)	Warm season	–	–
Guo et al., 2013	2007	China	Homes (59)	Warm season	GC-MS	–
Jia et al., 2008	2004, 2005	USA	Homes (159)	Summer, winter, fall and spring	GC-MS	Seasonal effects on indoor levels were inconsistent
Jo et al., 2003	2011	Korea	Homes (86)	Spring	GC-FID	–
Jo et al., 2004	2000	Korea	Homes (443)	Cold season	GC-FID	–
Kinney et al., 2002	1999	USA	Homes (46)	Winter and summer	GC-MS	There was a tendency for I/O ratios to be lower (closer to 1) in summer than in winter
Klinmalee et al., 2009	2005, 2006	Thailand	Schools (1)	Cold season	GC-FID	–
Kumar et al., 2014a	2011-2012	India	Library (1)	Winter and summer	GC-FID	Mean concentrations of TVOC, toluene were higher in winter
Kumar et al., 2014b	2011	India	Homes (27) Hostels (45)	Warm season	GC-FID	–
Larroque et al., 2006	2015	France	Schools (2)	Warm season	GC-MS	–
Lee et al., 2001	2000	Hong Kong	Restaurants (4)	Warm season	GC-MS	–
Lee et al., 2002b	1999	Hong Kong	Homes (6)	Warm season	GC-MS	–
Lu et al., 2006	2004	China	Hospitals (4)	Warm season	GC-MSD	–
Ohura et al., 2006	2001	Japan	Homes (46)	Winter and summer	GC-ECD	The concentrations of aromatic hydrocarbons and volatile organic halogenated compounds substantially increased in winter

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Table 2. (Continued).

Reference	Year	Country	Indoor environment (n)	Season	Analytical method	Seasonal variation
Ohura et al., 2009	2006, 2007	Japan and China	Homes (57 in Japan, 14 in China)	Summer and winter	GC-MSD GC-FID	In Japan, the concentrations of VOCs indoors were higher in winter. In China, higher concentrations were observed in summer.
Pegas et al., 2011a	2009	Portugal	Schools (14)	Warm season	GC-FID	–
Pegas et al., 2011b	2009, 2010	Portugal	Schools (14)	Winter, spring, autumn	GC-FID HPLC	For VOC, highest concentrations occurred during the coldest months. Formaldehyde concentrations in spring were higher than those in colder months.
Pekey and Arslanbas, 2008	2006, 2007	Turkey	Homes (15), schools (3), offices (10)	Winter and summer	GC-FID	There is a general increase of nearly all the compounds measured from summer to winter. For nearly all compounds, indoor VOC concentrations were significantly greater in winter than in summer.
Rios et al., 2009	2003	Brazil	Offices (2 buildings)	Winter	GC-FID, GC-MS	–
Roda et al., 2011	2003	France	Schools (28)	Cold season and hot season	GC-MS	No seasonal differences was identified
Sakai et al., 2004	1998	Japan and Sweden	Homes (37 in Japan, 27 in Sweden)	Winter	GC-ECD	–
Santarsiero et al., 2009	–	Italy	Dental clinic (1)	Warm season	GC-FID HPLC	–
Sarkhosh et al., 2012	2010, 2011	Iran	Photocopy centres (4)	Winter and spring	GC-MSD	The indoor VOC concentrations were higher in winter than in spring
Sax et al., 2004	1999 and 2000	USA	Homes (87)	Winter, summer and fall	GC-MSD	Mean concentrations of acetaldehyde, hexaldehyde, propionaldehyde, styrene, toluene, trichloroethylene, benzene, ethylbenzene, o-xylene, m/p-xylene, tetrachloroethylene, 1,1,1-trichloroethane were significantly higher in winter. Mean concentrations of formaldehyde were significantly higher in summer.
Sofuooglu et al., 2011		Turkey	Schools (3)	Fall, winter and spring	GC-MS	–
Son et al., 2003	2001	Korea	Homes (60)	Warm season	GC-MS	–
Tang et al., 2005	2002	China	Shopping malls (1)	Winter	GC-MSD	–
Tovalin-Ahumada and Whitehead, 2007	2002	Mexico	Office (33)	Warm season	GC-MS	–
Uchiyama et al., 2015	2011-2014	Japan	Homes (602)	Winter and summer	GC-MS	Almost all compounds were present at higher levels in summer than in winter
Ullrich et al., 2002	2001-2002	Germany	Schools (44)	Winter	GC-MS	–
Zhou et al., 2011	2008	China	Homes (10) Offices (6)	Summer	GC-MSD	–

GC-MS: Gas chromatography coupled with mass spectrometry; GC-FID: Gas chromatography coupled with flame ionization detector; GC-MSD: Gas chromatography coupled with mass selective detector; GC-ECD: Gas chromatography coupled with an electron capture detector; VOC: total volatile organic compounds;

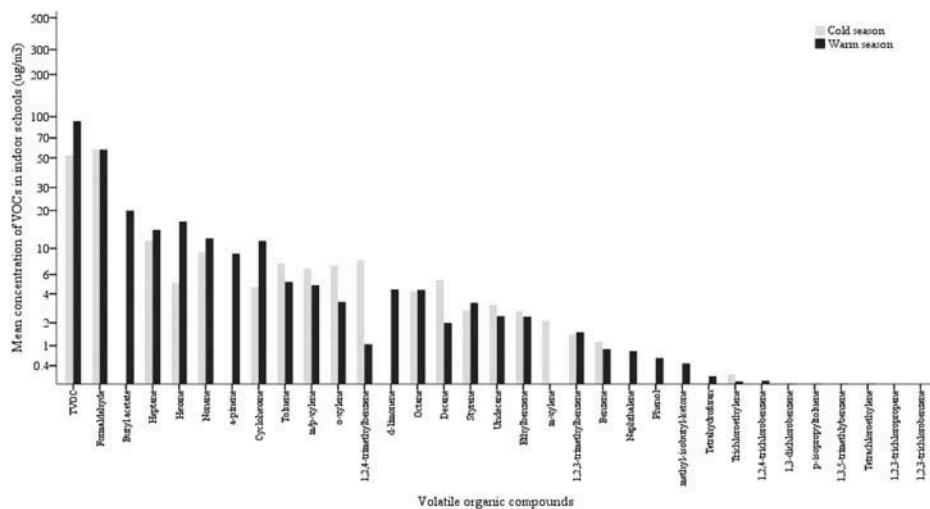


Figure 2. Comparison of VOCs mean concentrations measured in schools in cold and warm season.

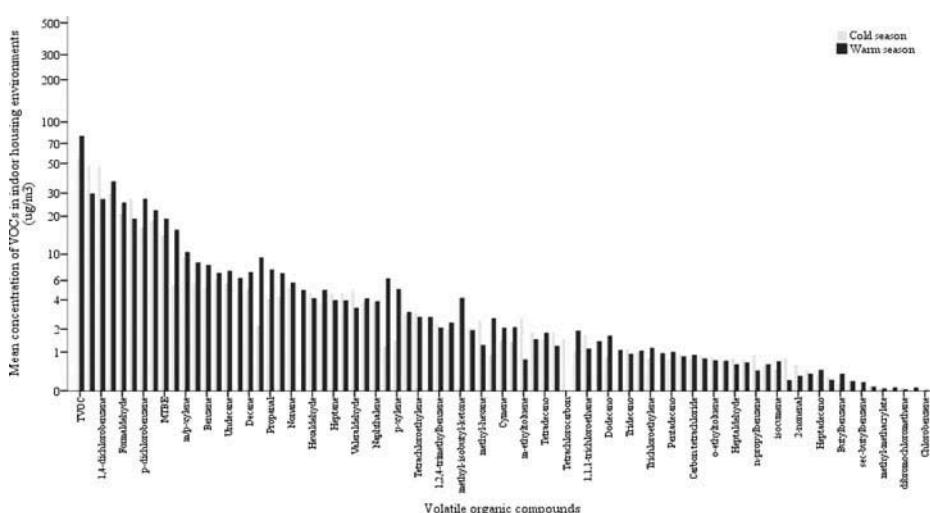


Figure 3. Comparison of VOCs mean concentrations measured in housing environments in cold season and warm season.

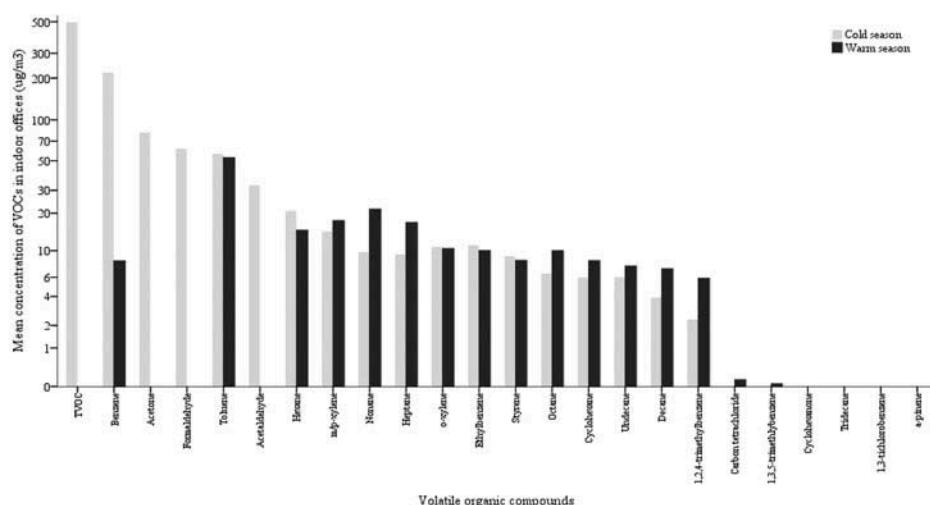


Figure 4. Comparison of VOCs mean concentrations measured in offices in cold season and warm season.

concentrations of most specific VOC, with measurements in both seasons, were higher in the warm season (**Figure 4**). The levels ranged between $0.05 \mu\text{g}/\text{m}^3$ and $532 \mu\text{g}/\text{m}^3$, higher for benzene in the cold ($404 \mu\text{g}/\text{m}^3$) and lower for 1,2,3-trimethylbenzene in the warm season (supplementary file).

In other indoor environments, including commercial spaces, hospitals, restaurants, dental settings, photocopy centers, and museums considered in the analysis, 26 VOC were measured and reported, with 11 present in both seasons, 13 in the cold and only 2 in the warm season (benzaldehyde and carbon tetrachloride). For most VOC the concentration was higher in the cold compared with the warm season (**Figure 5**). The levels ranged between $7.32 \times 10^{-4} \mu\text{g}/\text{m}^3$ and $185.1 \mu\text{g}/\text{m}^3$, which were observed in the cold season (benzene and toluene, respectively) (supplementary file).

There were six VOC common to the four previously described indoor environments and determined in both seasons. The highest concentrations were observed in cold season and in offices (benzene [$334 \mu\text{g}/\text{m}^3$], *o*-xylene [$10.8 \mu\text{g}/\text{m}^3$], and styrene [$9 \mu\text{g}/\text{m}^3$]), in other indoor environments (*m/p*-xylene [$38 \mu\text{g}/\text{m}^3$]), and in housing environments (toluene [$171 \mu\text{g}/\text{m}^3$]) (**Figures 2–5**). Only for ethylbenzene were there found higher concentrations in the warm season ($20 \mu\text{g}/\text{m}^3$) in offices. When comparing all individual VOC for each different indoor environment, no marked differences were noted between VOC concentrations and seasons. When

considering the VOC that were common to the four indoor environments (schools, housing, offices, and other indoor), significant differences were also not observed for the seasons.

The geographical assessment was analyzed by dividing data into continents: Europe, Asia, and America. In the cold season, higher mean concentrations of benzene indoors were found in offices in America; for the remaining five VOC higher mean concentrations indoors were observed in Asia (ethylbenzene, toluene, and *m/p*-xylene in other indoor environments, *o*-xylene in offices, and styrene in housing). In the warm season, for most indoor VOC (benzene, ethylbenzene, toluene, and *m/p*-xylene, and styrene) higher mean concentrations were detected in Asian schools; for *o*-xylene higher mean concentrations were found in Asian offices. However, no significant differences were observed for the continents (Figures 1S and 2S, supplementary file).

Seasonal variations in indoor/outdoor ratio

The indoor/outdoor (I/O) ratio was estimated for each environment by season (**Figures 6–9**) in order to determine the origin of possible VOC sources. In schools there were 31 measured compounds common to both indoor and outdoor environments and the values ranged between 0.51 (1,2,4-trimethylbenzene) and 23 in the cold season (methyl-isobutyl-ketone). Based on 17 specific VOC and comparing cold and warm seasons, the I/O ratio was higher than unity for the majority of

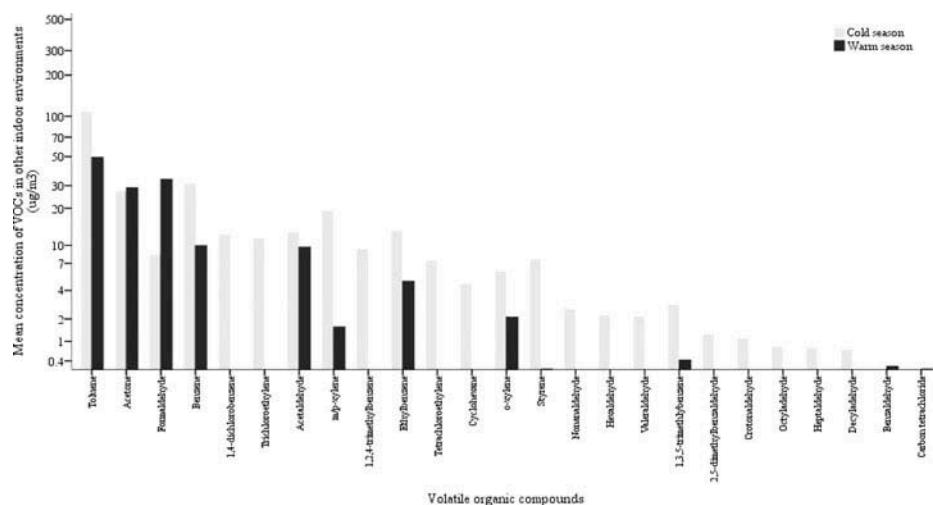


Figure 5. Comparison of VOCs mean concentrations measured in other indoor environments in cold season and warm season.

VOC in both seasons (10 of 17), but mostly higher in the cold season; only 3 VOC (benzene, trichloroethylene and hexane) displayed values lower than 1. For the remaining VOC with information just for one season (1 in the cold and 13 in the warm season) the I/O values ranged from 1 (1,2,3-trichlorobenzene and 1,2,3-trichloropropane) to 23 (methylisobutyl ketone) (Figure 6).

In the housing environments a greater number of VOC (71) with both indoor and outdoor concentrations were identified. There were in total 61 VOC reported in both seasons, and for most of these the I/O was higher than 1 (57) and higher in the warm season. Four VOC presented in one of the seasons an I/O ratio lower than 1 (*o*-xylene, tetrachloroethylene, and *p*-xylene in the warm

season [0.93, 0.91, and 0.31, respectively] and carbon tetrachloride in the cold season [0.87]). For VOC identified in only one season (10), 9 of them presented an I/O higher than 1 (Figure 7).

There were 19 measured VOC common to both indoor and outdoor office environments. Fourteen of them had values of I/O for both seasons and mostly higher than 1 (11 compounds all in the warm season). Only four VOC presented I/O values for warm or cold seasons, of which two had an I/O higher indoors (Figure 8).

In other indoor environments (Figure 9) and for VOC with I/O for both seasons (8), only 2 presented values lower than 1 (formaldehyde in the cold season [0.79] and *p*-xylene in the warm season [0.76]). Higher I/O values were observed in the cold season

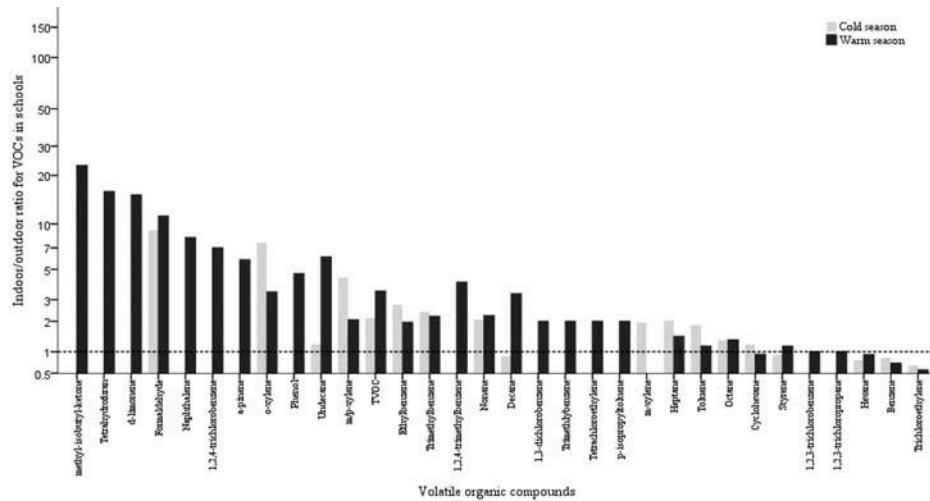


Figure 6. Comparison of indoor/outdoor ratio of VOCs mean concentrations measured in schools in cold season and warm season.

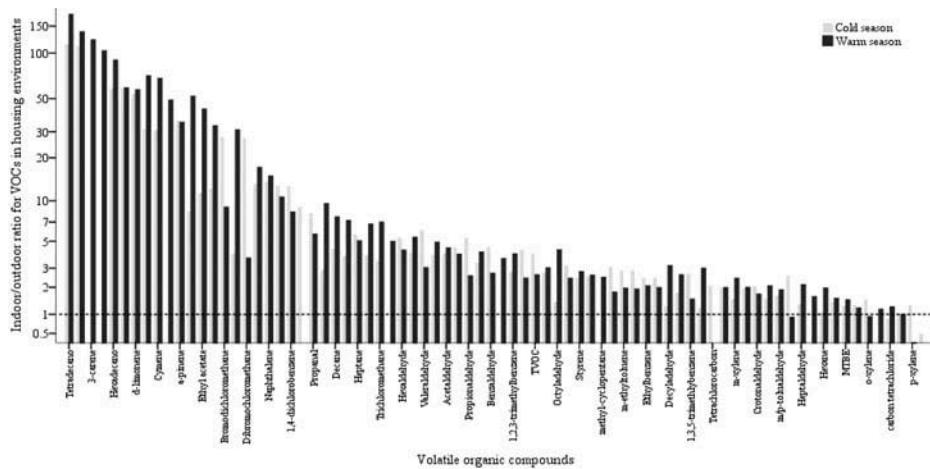


Figure 7. Comparison of indoor/outdoor ratio of VOCs mean concentrations measured in housing environments in cold season and warm season.

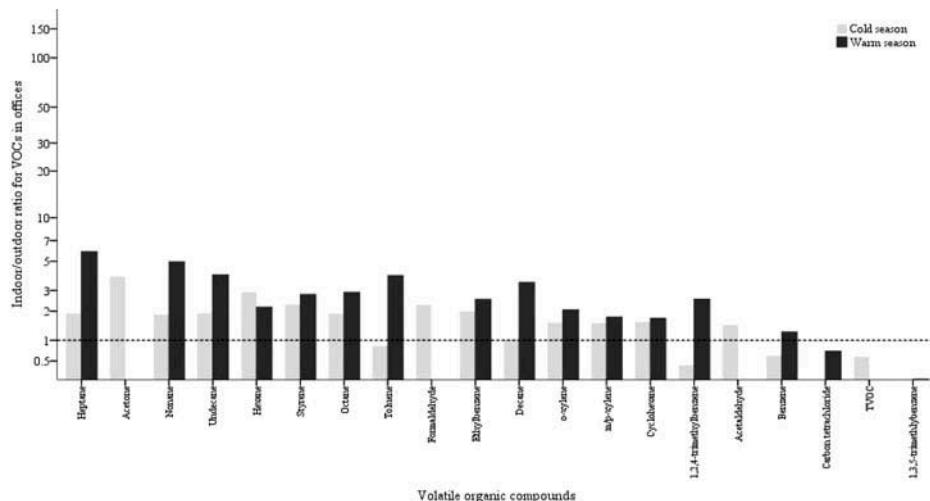


Figure 8. Comparison of indoor/outdoor ratio of VOCs mean concentrations measured in offices in cold season and warm season.

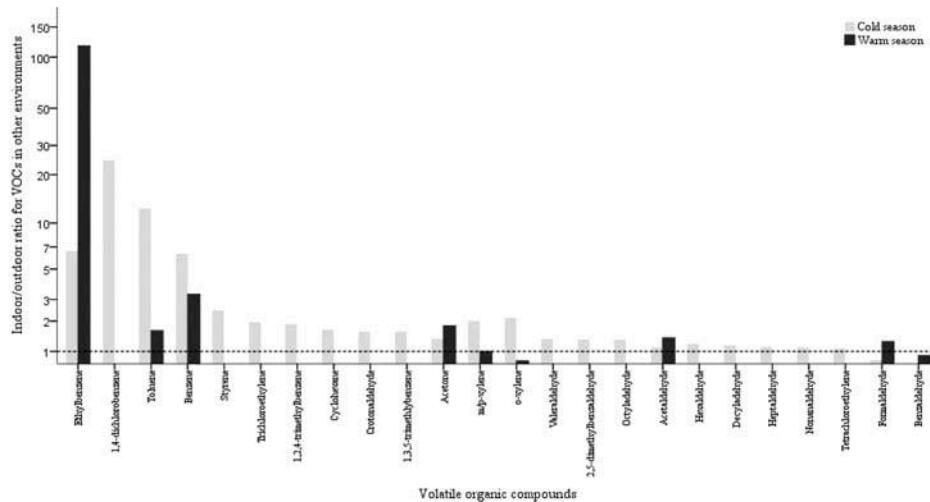


Figure 9. Comparison of indoor/outdoor ratio of VOCs mean concentrations measured in other indoor environments in cold season and warm season.

for most VOC. From the VOC with measurements in a single season (16 compounds) only I/O for formaldehyde was lower than 1 (0.89 in the warm season).

In housing environments, unlike the other three indoor environments, there was a significant difference between VOC levels indoors and outdoors considering the analysis by season (Figure 7). Concentrations of formaldehyde, 1,2,3-trimethylbenzene, and acetaldehyde were significantly higher indoors than outdoors for both seasons. Indoor concentrations of ethylbenzene and 1,3,5-trimethylbenzene were significantly higher only in the cold season. Higher concentrations of toluene, styrene, heptane, nonane, decane, and 1,2,4-trimethylbenzene were also observed indoors, but significantly elevated only in the warm season.

In the cold season, highest levels for outdoors were observed in Asia (styrene in schools, ethylbenzene and *o*-xylene in offices, and *m/p*-xylene in other environments) and America (benzene and toluene in offices); in the warm season, higher concentrations for all six VOC were observed in Asian schools (Figures 3S and 4S, supplementary file). However, no marked differences were noted for the continents for both seasons.

Discussion

This review summarized and compared existing data on type and levels of individual VOC detected, measured, and reported both indoors and outdoors by season in four major indoor environments (schools,

housing, offices, and other indoor). As observed, VOC are commonly found in indoor air, and the specific compounds and their concentrations vary between indoor environments and season. In general, the collected data indicated that levels of indoor VOC were higher in housing environments and offices. A higher number of compounds were also identified at housing environments. This might be due to several indoor sources in both environments, and activities and behaviors of occupants in the respective buildings, especially in housing environments. Unlike offices where occupational exposure limits need to be respected, no regulatory limit or public health objective has been set for residential indoor spaces, which may also contribute to higher VOC concentrations. According to Symanski et al. (2009), housing environments are affected by one or more potential sources of VOC, including the use of consumer products (household cleaners, air freshness, deodorizers, personal care products, waxes, and pesticides), tobacco smoke, cooking, and heating appliances.

The VOC identified in each environment, especially indoors, and their concentrations varied considerably among the studies. This variation may be attributed to differences in sampling methods and geographic settings, as well as fluctuations in air exchange rates, occupants' behaviors, and outdoor air concentrations. The large range of levels among the studies reviewed supports the use of concentration distributions rather than a single measure of distribution to characterize concentrations in indoor air.

In the four indoor environments considered in the current review, the mean concentration of most of these compounds was higher in the cold season, showing that there is a time variation of VOC composition and level during different seasons and providing information regarding the influence on indoor air quality from occupants and their behaviors and from climatic conditions. It might be due to lower ventilation, because during the cold season, most of the time windows or doors are closed, and lower air exchange rates contribute to a gradual accumulation of VOC (De Blas et al. 2012; Madureira et al. 2016).

As observed, geographical differences (like local sources, culturally related human behavior, and climate) may affect the concentrations of VOC outdoors and indoors. Although no significant

differences were found between the VOC concentrations by continents and for both seasons, weather patterns in each region/country may exert an important impact on the outdoor and indoor concentrations, due to changing human behavior related to weather circumstances, opening windows during warm or cold days, and heating habits (e.g., wood vs. gas heating) (Ballesta et al. 2006; Ilgen et al. 2001). Sarigiannis et al. (2011) also reported a variation in VOC concentrations indoors in different geographical locations in Europe. Sarigiannis et al. (2011) found higher concentrations of formaldehyde in northern Europe than in the south, especially in homes, which was explained by different ventilation schemes between north and south, due to the differing climatic conditions, resulting in lower air exchange between indoor and outdoor environment, as well as with furniture and building materials in the European north. For benzene and toluene the same study demonstrated a greater influence of outdoor sources (such as transport) on indoor air concentrations in the south than in the north, because of the higher outdoor-to-indoor air penetration observed due to the different climates (warmer in the south).

The I/O ratios were larger than 1 for nearly all VOC identified in different environments, suggesting that indoor sources and poor ventilation provide a greater contribution to the levels of VOC indoors. However, for some VOC, an I/O close to or below 1 was found, indicating that indoor concentrations were affected by outdoor air. Outdoor emission sources, such as traffic and industries (which strongly depend on the degree of urbanization), and outdoor-to-indoor air penetration also supply an important contribution to VOC and levels observed indoors.

The outdoors VOC concentrations for the different continents may be explained, in addition to different traffic patterns and industries, by meteorological conditions. For Europe, Cocheo et al. (2000) noted an increase in annual average outdoor VOC concentrations from north to south, which was attributed to differences in prevailing meteorological conditions, such as local wind speeds. Lower wind speeds have been associated with less dispersion and higher outdoor concentrations in the region (Miller, Xu, and Luginaah 2009; Sari et al. 2014).



Some interventions need to be made to reduce indoor VOC concentrations and consequently the risks of exposure, such as avoiding redecoration, new furniture, air fresheners, and perfumed items, and increasing ventilation (Annesi-Maesano et al. 2013; Dales et al. 2004) and banning tobacco smoke at home (Brajenovic, Karaconji, and Bulog 2015). Low-VOC-emission materials and consumer products, decrease in number of students in classrooms, and avoiding use of air cleaners, incense, and candles (Lim et al. 2014; Pegas et al. 2011b) might also be considered as interventions in the different indoor environments. Further studies are also needed to evaluate the effectiveness of interventions to reduce the VOC concentrations.

The main limitations of this review are (i) number of assessed individual VOC; (ii) distribution of samples collected in each environment; (iii) amount of studies that reported both indoor and outdoor VOC concentrations in various seasons; and (iv) different methodologies used for sampling VOC. A further limitation of our review is the combined analysis of the results without considering tobacco smoke exposure. However, and regarding housing, offices, and other indoor environments, few studies presented results by exposure to tobacco smoke and some did not report whether the occupants smoked indoors.

The overview of the results indicated that for some VOC a wide variability is observed, due to the different source characteristics. Thus, their presence and concentrations are limited to each indoor environment, season, and geographical location, where socioeconomic, regulatory, or consumer behavior greatly affects indoor air quality. For some VOC, strong outdoor emission sources and outdoor-to-indoor air penetration might have a more significant contribution than indoor sources, making source apportionment even more complicated (Sarigiannis et al. 2011).

This review provides an overview of VOC concentrations and compounds in 4 major indoor environments (schools, housing, offices and other indoor) over the last 20 years. Comparisons of results by season and ratios I/O support the importance of sampling VOC throughout the year and also confirm that indoor sources (in both seasons) play a major role in the exposure to VOC.

Conclusions

An overview of the results indicated that for some contaminants a wide variation was observed due to different source characteristics. This review confirmed that most VOC identified indoors are linked to indoor sources and to specific activities and behaviors of the occupants. A seasonal cycle in VOC concentration and compounds was also identified in all indoor environments. Given the number and range of VOC concentrations observed indoors, it is important to assess indoor air pollutants, namely, VOC, in order to determine the potentially attributable health risk and to identify the most appropriate risk management strategies. In this context it is important to consider sensitive subpopulations when assessing risk characterization and hazard (Valcke and Haddad 2015). It is noteworthy that actions and policies to reduce VOC exposures, such as improved product labeling and consumer education, are recommended.

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ORCID

- Joana Madureira <http://orcid.org/0000-0001-5279-877X>
 João Rufo <http://orcid.org/0000-0003-1175-242X>

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